

**i. Cover Page**

PROPOSAL  
to  
COMPUTATIONAL MATERIALS SCIENCE NETWORK  
of the  
Department of Energy Basic Energy Sciences  
COOPERATIVE RESEARCH TEAM  
on  
**MULTISCALE STUDIES OF THE FORMATION AND  
STABILITY OF SURFACE-BASED NANOSTRUCTURES**

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## **1. CRT Title and Executive Summary**

# **MULTISCALE STUDIES OF THE FORMATION AND STABILITY OF SURFACE-BASED NANOSTRUCTURES**

## **Executive Summary**

In this proposal, a team of distinguished researchers with highly complementary expertise is assembled to carry out multiscale studies of the formation, stability, and novel physical properties of important classes of surface-based nanostructures: nanoclusters and quantum dots (zero-dimensional, or 0D), quantum wires and quantum wire superlattices (1D) and ultrathin quantum films and platelets (2D). As is widely recognized, the ability to precisely control the formation of innovative nanostructures of technological significance, as well as to preserve their integrity under diverse practical conditions, is a grand challenge in nanoscience and nanotechnology. In particular, ordered arrays of quantum dots, quantum wires, and quantum wire superlattices of alternating magnetic and nonmagnetic (or insulating) elements are among the most desirable artificially-structured nanosystems of the experimental community, owing to their huge potential as elemental building blocks in future device applications. Our primary objective is to make major conceptual advances in growth science, characterized by fundamental understanding and accurate prediction of the evolution of the prototype nanostructures. This objective is to be achieved through collaborative computational efforts and development of new mathematical tools and algorithms to provide a coherent study of the problems from the electronic and atomistic to the continuum levels. Such advances in better structural control will not only facilitate more reliable property studies of such low-dimensional nanostructures, but will also enable direct comparison with experiments. The multiscale models and computational methods to be developed through the integrated efforts of the cooperative research team (CRT) will be optimized for application in other important areas of nanoscience as well.

## **2. Overview of Proposal and Management Plan**

### **2.1 Overview of Proposal**

#### **2.1.1 How will science be advanced?**

In the rapidly expanding field of nanoscale science and technology, one widely recognized bottleneck issue is the fabrication of desirable nanostructures with atomic-scale precision. Important example systems include group IV [1,2] and III-V [3] semiconductor quantum dots for optical and nanoelectronic applications, and magnetic/metal nanoclusters [4,5] for memory and nanoscale catalytic devices. Here it is often highly desirable to have large arrays of nanoclusters with narrow size and uniform spatial distributions, yet to date proven powerful approaches to meet such needs of the scientific community are still lacking. Another important class of systems is metallic and/or magnetic quantum wires [6,7] and quantum wire superlattices on various substrates [8]. Here again, precise control of their spatial arrangements, both in “straightness” and in chemical composition, is still far beyond reach.

The bottleneck problem in fabrication poses a major obstacle in fundamental and applied studies of surface-based nanostructures, both on the experimental and theoretical side. On the experimental side, without ideal testing grounds fundamental physical laws in systems of reduced dimensionality, such as breakdown of Fermi liquid theory in one-dimensional electrical transport, cannot be tested unambiguously. On the theory side, lack of knowledge of the precise atomic arrangements often forces researchers to invoke unwarranted assumptions when exploring the intriguing physical properties and potential technological applications of such nanostructures.

Over the years, a great deal of experimental and theoretical research effort has been devoted to the bottleneck problem of fabrication, and a wealth of conceptual advances have been made [6,9,10]. Here we propose to take full advantage of recent progress in this vitally important field by assembling and integrating two teams of researchers active in the field, one in experiment and one in theory, to work collaboratively to explore innovative approaches for the formation of a few well-selected “dream” systems of the community, such as highly ordered quantum dot arrays, quantum films, quantum wire superlattices, and magnetic nanoplatelets. We will pay particular attention to controlled fabrication of magnetic nanostructures on silicon substrates, with the technological objective of integrating ultrahigh density information storage and information processing on single chips.

Because most such technologically relevant artificial nanostructures at surfaces are not in thermodynamically stable configurations, we must “trick” nature by devising kinetic pathways to reach such metastable configurations by exploiting the delicate interplay between kinetic and thermodynamic factors. The acquisition of accurate information on growth kinetics as characterized by various atomic rate processes necessarily demands first-principles computational efforts at the electronic and atomic scales, while proper treatment of thermodynamic driving forces such as the stress field associated with the lattice mismatch in a heteroepitaxial growth system demands theoretical approaches spanning length scales from the atomic to mesoscopic. The latter length scale is far beyond the scope of the state-of-the-art first-principles approaches, and has to be tackled using continuum techniques built on, for example, elasticity theory.

Besides formation, the intrinsic thermodynamic stability (or, more precisely, the *metastability*) of the nanostructures is another fundamentally important aspect of our research emphasis [11-13]. Knowledge about such metastability not only serves as guidance in selecting physically accessible nanostructures in the formation process, but is also indispensable in exploring the application potentials of the nanostructures.

### **2.1.2 Who will care and why?**

A unique feature of this CRT is the truly substantial integration of a network of experimental research teams into the proposed theoretical effort (see 2.2.1). Many of the theoretical issues to be addressed in this proposal, in particular the selection of the prototype model systems, were formulated on the basis of the inputs and challenges presented to us from the experimental side during the first workshop of this research effort (Emory University, January 18-19, 2003) and continued close interactions till today. In this regard, it is fair to state that the proposed theoretical program is a direct and much needed response to the experimental scientific community. The composition of the experimental team, including major research groups at three national labs (Ames, ORNL, and Sandia) and three NSF-sponsored research centers at universities (Nanoscale Science and Engineering Center at Harvard, and Materials

Research Science and Engineering Centers at Maryland and at Wisconsin), also represents well the experimental community. The objectives of this CRT correspond very well with the thrusts of these federally funded leading research centers.

Given the lack of direct funding allocation from the CMSN project to the experimental side, one might wonder why the experimental groups would care to participate in the proposed research collaborations. The reason, of course, lies in scientific motivation, as evidenced by past track records of substantial and fruitful collaborations between various members of the experimental and theoretical teams. One naturally expects that the integrated program proposed here will dramatically enhance the effectiveness of collaborative research efforts in addressing some of the major challenging problems faced by the community as a whole, including both theorists and experimentalists.

It should be noted that, because the present CRT focuses on the enabling step of formation of important classes of nanostructures, advances in this CRT should naturally draw the interests of other potential CRTs focusing on addressing various physical properties of such nanostructures and related low-dimensional systems. In this regard, we anticipate that the present CRT will help to facilitate substantial inter-CRT collaborations as well.

Ultimately, major advances in fundamental understanding and physical realization of the formation and stability of technically significant nanostructures will undoubtedly be beneficial to all working in the fields of nanoscale science, engineering, and technology.

### **2.1.3 The rationale for a team effort**

It is encouraging that recent substantial collaborations between the theorists and between theory and experiment have been fruitful at several localities and also between different institutions. For theoretical collaborations, examples include Ho, Wang, and Shenoy on semiconductor quantum dots [14,15], and Kaxiras and Zhang on low-dimensional magnetic nanostructures [16,17]. For collaborations between theory and experiment, local examples include Einstein (theory) and Williams (experiment) at Maryland [11,18,19], and Ho, Wang and Evans (theory) and Tringides and Thiel (experiment) at Ames [20-24]; cross-institution examples include Chou at Georgia Tech (theory) and Chiang at Illinois (experiment) [25,26], and Zhang at ORNL/UT (theory) and Lagally of Wisconsin and Shih at UT-Austin (experiment) [13,27-29]. Nevertheless, these existing collaborations are still limited in their scope of research emphasis and expertise. The proposed CRT program will integrate the complementary strengths of the participating groups to tackle the big questions that would otherwise be impossible for any individual research effort to address. In particular, the multitude of scientific issues involved and the corresponding wide variety of theoretical approaches required at different scales, ranging from first-principles electronic calculations to atomistic molecular dynamics and kinetic Monte Carlo simulations to mesoscopic scale modeling, call for a comprehensive team effort as proposed here.

### **2.1.4 The expected impact**

The present proposal brings together a comprehensive team of experts in theoretical modeling and simulation of key problems in the formation and stability of surface-based quantum structures from the electronic and atomistic up to mesoscopic length scales. Through the study of prototype systems motivated by the experimental community, these multiscale simulations are expected to advance our fundamental understanding of the key issues governing the formation and integrity of surface structures at the nanoscale. Collaboration between groups

with complementary expertise will eventually lead to the development of multiscale simulation packages which can be used for much more accurate prediction of material properties on surfaces guiding the way to “dream” structures pursued by experimental groups. These advances will have an important impact on a broad class of nanodevices such as the integration of nanostructures with silicon chip technology. In particular, we expect to devise multiple new kinetic pathways for fabricating low-dimensional metallic and magnetic nanostructures on silicon by fully exploring the power and strength of quantum mechanical driving forces at the nanoscale.

## 2.2 Management Plan

### 2.2.1 CRT members

This cooperative research team (CRT) consists of three sets of researchers as given below. The first set, listed on the cover page, includes the team coordinators (Ho and Zhang) and the six task leaders (Chou, Einstein, Evans, Kaxiras, Shenoy, and Wang). These are the core members of the CRT who are fully committed to execute the research activities spelled out in Section 4.2. The second set of members includes Family, Feibelman, Liu, Suo, and Tersoff. Each of these five theorists has expressed an interest to participate in the general research activities of the present CMSN project, including the annual coordination meetings. Collectively, these five theorists will add complementary strength and vision to the CMSN program. Together, these two sets of theorists encompass a broad spectrum of computational expertise ranging from electronic and atomistic calculations to continuum modeling and simulations. The third set consists of the experimentalists, who will present challenging problems from the experimental community to the team and also provide critical assessment of the validity of the theoretical approaches being developed. Most of the participants have DOE and/or NSF funded research programs related to nanostructure formation and stability. Collaborations have existed among the team participants on various individual research projects. The effort of this CRT will enhance such collaborations in a more coherent and efficient way in order to be able to tackle the grand challenge issues discussed above. Participants in the CRT are from three DOE laboratories (Ames, ORNL, and Sandia), three NSF Materials Research Centers (Harvard, Maryland, and Wisconsin), eleven universities, and one company (IBM). The education and research backgrounds of the core CRT members are listed in the two-page CV section (pages 30-45).

#### Core members

##### **Team Coordinators:**

Kai-Ming Ho

Ames Lab/Iowa State Univ.

Zhenyu Zhang

Oak Ridge National Lab/Univ. of Tennessee

##### **Task Leaders:**

Mei-Yin Chou

Georgia Institute of Technology

Theodore Einstein

University of Maryland

James Evans

Ames Laboratory/Iowa State University

Efthimios Kaxiras

Harvard University

Vivek Shenoy

Brown University

Cai-Zhuang Wang

Ames Laboratory

#### Participants

##### **Theory:**

Fereydoon Family  
 Peter Feibelman  
 Feng Liu  
 Zhigang Suo  
 Jerry Tersoff

Emory University  
 Sandia National Labs  
 University of Utah  
 Harvard University  
 IBM

#### **Experiment:**

Michael Aziz  
 Tai-Chiang Chiang  
 Gary Kellogg  
 Max Lagally  
 Jian Shen  
 Chi-Kang Shih  
 Brian Swartzentruber  
 Patricia Thiel  
 Michael Tringides  
 Hanno H. Weitering  
 Ellen Williams

Harvard University  
 UIUC  
 Sandia National Lab  
 University of Wisconsin  
 ORNL  
 University of Texas  
 Sandia National Labs  
 Ames Lab/Iowa State Univ.  
 Ames Lab/Iowa State Univ.  
 ORNL/Univ. of Tennessee  
 University of Maryland

### **2.2.2 Meetings**

We will plan annual CRT coordination meetings (two-days) to discuss research progress and research plans for the following year. Selected outside experts will be invited to the meetings. Together with students and postdocs, participation at such meetings could involve as many as 40 scientists. Travels and extended visits of postdocs and team members to other participating groups and small sub-task meetings will be planned to promote collaborations between distant groups. We will make use of Internet communication and conferencing facilities whenever possible, and will create a set of project web pages where preprints, updates, and comments can be shared.

### **2.2.3 Coordination**

The coordinators responsible for the overall project are **Kai-Ming Ho** (Ames Lab) and **Zhenyu Zhang** (ORNL). Each task will also be coordinated by a task leader.

## **3. Budget and Budget Justification**

In order to establish strong partnerships among members of the CRT, the CMSN fund will be used to support eight co-shared postdocs or graduate students (50% from CMSN funding and another 50% matched from the team members' existing research grants and some matching funds from the experimental side (MRSECs)) to work collectively on the different aspects of the research projects as described in Sec. 4.2. Funds are also allocated for the annual coordination meetings. The budget for FY 2005 (with full overhead) is:

50% of 8 postdocs or students	\$250K/year
Annual coordination meeting	\$20K/year
Web pages/misc.	\$10K/year
<b>Total</b>	<b>\$280K/year</b>

## 4. Narrative

### 4.1 Background and significance of CRT and preliminary studies

This proposal addresses two key aspects in the realization of nanostructured devices (e.g. quantum dots, quantum films, quantum wires, and magnetic nanoplatelets) on surfaces. One is how to form such well-defined nanostructures in designated or patterned locations on surfaces. The other is to explore the stability of such nanostructures once they are formed. The scientific issues to be addressed in the present proposal can be grouped into five main areas as follows, each demanding the use of existing as well as new computational approaches spanning more than one length scale. The five areas are not only intimately connected scientifically, but also they must be modeled concertedly over all the length scales from electronic and atomic to the continuum.

#### 4.1.1 Elemental atomic rate processes

To form a feature nanometer in size and with well-defined shape, atoms deposited onto a nominal surface must be able to diffuse across the terrace and reach each other to nucleate and grow. This seemingly simple process actually demands an exceptionally high degree of concerted motion of many adatoms. Even in the case of adatom motion on a flat terrace, the preferred diffusion pathway can involve the concerted motion of both the diffusing adatom and one or more surface atoms [30]. Furthermore, an adatom may encounter a higher potential energy barrier as it attempts to go across a step edge [31,32]. Figure 1(a) illustrates the various important atomic processes involved in the fabrication of surface features such as faceted nanoclusters or in thin film growth, emphasizing primarily downward motion of adatoms [27]. In Figure 1(b), a concerted atomic process for upward motion of adatoms at a step edge on a metal fcc(110) surface is illustrated, whose crucial importance in nanocrystal formation and faceting has been established using first-principles calculations within density functional theory coupled with kinetic Monte Carlo growth simulations (*ab initio*-KMC). [33,34]

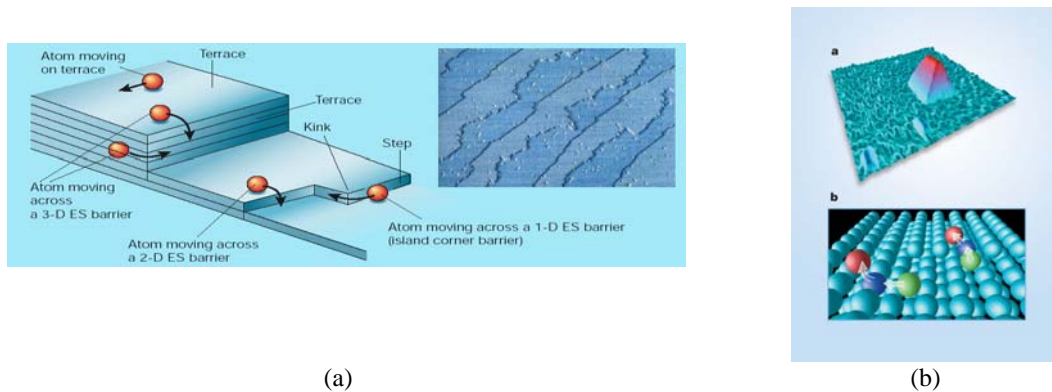


Fig. 1. (a) Illustration of different types of downward atomic diffusion processes [27]. (b) Illustration of upward atomic diffusion at single-atom-layer-high steps on Al(110), via a concerted place exchange mechanism [33,34].

Although first-principles total-energy calculations can be employed to determine accurately the energy barriers if the atomic pathways of the kinetic processes are known, it is too expensive in the foreseeable future to use first-principles methods to fully explore the complex

pathways of kinetic processes associated with nanostructure growth and evolution on surfaces. Efficient search engines need to be developed with accurate interatomic interactions so that the potential energy landscapes can be investigated in detail before applying first-principles approaches. Tight-binding molecular dynamics (TBMD) developed at Ames laboratory can serve ideally this purpose [35]. For a given system, the method is several thousand times faster than first-principles calculations, yet the interatomic interactions can be fitted to a reasonable accuracy to capture the essential kinetic processes on surfaces. The power of this hybrid DFT/TBMD method has been demonstrated recently in uncovering intriguing diffusion pathways of Si adatom and addimer on Si(100) [36,37]. Extension of this method to more complex situations such as steps, kinks, and the growth front in heteroepitaxy will be one of the major computational challenges in this program.

#### 4.1.2 Step energetics and dynamics

A realistic surface is never perfectly flat, but contains a variety of defects, with steps as the one most relevant for growth of nanostructures. New steps are generated when islands form on initially flat terraces. As line defects, steps themselves are commonly defective, containing kinks as they meander around some mean paths. An example is shown in the insert of Fig. 1(a), for the meandering of two different types of steps ( $S_A$  and  $S_B$ ) on a vicinal Si(100) surface [38].

For many practical purposes, one would desire the steps to be as straight as possible. This is the case for patterning a surface with monatomic-layer-high islands with regular shapes and uniform spatial distribution. It is even more so when pre-existing steps on a vicinal surface are to be used for fabrication of ordered arrays of quantum wires as illustrated in Fig. 2 (see Sec. 4.2.3) [6,7,39].

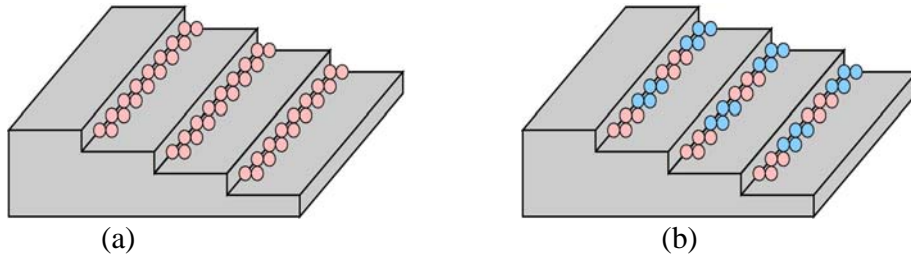


Fig. 2 Illustrations of (a) ordered wire arrays, and (b) quantum wire superlattices on vicinal substrates.

Qualitatively, the straightness of steps is determined by the step and kink formation energies. For a given system, the step formation energy and step-step interactions are dependent not only on the detailed chemical bonding at the steps, but also on the strain field created by the atomic reconstruction at the steps. To calculate such energies, one has to invoke accurate descriptions of physical interactions at two very different length scales: short-ranged electronic interactions near the steps [40] and long-ranged elastic interactions far away from the steps. Although there has been some progress in treating such multi-scale computational problems in other fields [41,42], efforts to determine step formation energy and step-step interactions from electronic and atomistic calculations are limited. Multiscale algorithms and methods need to be developed to tackle the problem accurately.

Over the years, researchers on this CRT have carried out pioneering studies of step energetics and dynamics on both Si(100) and (111) surfaces as well epilayers on such surfaces. [38,43-45] In particular, bunched steps are generally more nearly straight than any of the single

constituent steps, and may serve as better templates for ordering nanoclusters or fabrication of quantum wires or potential quantum wire superlattices. [6,7,39,46] One emphasis of this CMSN program will be to use the DFT/TBMD energy and rate parameters, coupled with a long-ranged strain description of step dynamics and step-step interactions, to search for optimal growth conditions for ordering steps with desired atomistic and compositional configurations. In this approach, the interplay of the Ehrlich-Schwoebel barrier effects [31] in step dynamics and step-step interactions will play an essential role as detailed in Sec. 4.2.

#### 4.1.3 Island nucleation and growth

In a standard thin film deposition experiment on a flat surface, monatomic-high islands and nanoclusters are formed with density and size distributions satisfying well-defined scaling laws within classical nucleation theory [47-49]. A great deal of conceptual understanding has been achieved in controlling dynamical island growth under various physically realistic growth conditions. Nevertheless, the islands formed are typically spatially disordered and have broad size distributions. To improve spatial ordering and size uniformity, the most promising route is self-assembly due to surface reconstruction on a patterned template [50,51]. For metal nanoclusters grown on semiconductor substrates, we have recently demonstrated that, when combining the power of magic clustering and self-assembly via optimal growth control, spatially ordered identical metal and magnetic nanoclusters can be formed over large areas on a Si(111)-7x7 reconstructed surface [4,52]. The formation of such ideal 2D nanocluster arrays can be described in terms of different adsorption and diffusion behaviors of deposited metal atoms on the faulted or unfaulted half unit cell of the Si(111)-7x7 surface, an idea originating from previous theoretical studies [53]. Such ideally ordering nanoclusters offer huge potentials for developing nanodevices with chemical, magnetic, and optical applications [54-57]. As will be shown in Section 4.2.3, such ordered 2D nanocluster arrays may further be exploited as growth templates for quantum engineering of magnetic nanoplatelets and other complex nanostructures with technological significance [29].

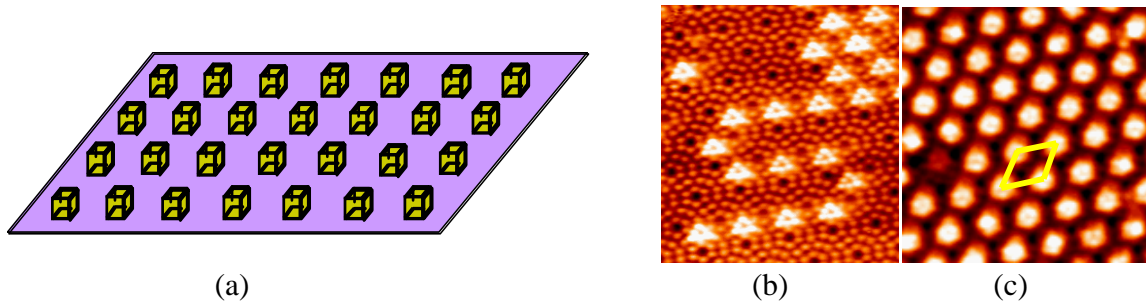


Fig. 3. (a) Illustration of one “dream” type of surface nanostructure: an array of nanoclusters with identical size and uniform spatial distribution. In nanoclusters formed on Si(111)-7x7 at low coverage (b) and high coverage (c) as revealed by the STM, with (c) essentially a physical realization of the dream nanostructure in (a) [4]. Each white spot in (b) and (c) represents a magic cluster of 6 In atoms.

Clearly, a complete understanding of the formation mechanism of the self-assembly of large numbers of nanoclusters on a substrates with reconstructed superstructures is well beyond the scope of full quantum mechanical description based on DFT calculations. For properly chosen prototype model systems, TBMD can again serve to increase the length scale to thousands of atoms, but the time scale for self-assembly is still a big problem. Here, the use of *ab*

*initio*-KMC will likely prove to be essential in identifying some of the most important atomic rate processes in the self-assembly.

When clusters grow into the multilayer regime, the atoms in the upper layers will feel much weaker effects from the underlying substrate, unless the islands are coherently strained. How the island morphology evolves will depend on the various kinetic factors [33], strain energy, and, for metal nanoclusters on semiconductor substrates, the driving force associated with the confined motion of the conduction electrons within the metal islands [58,59]. Such considerations will further push the length scale of the system to larger values, and because of the need to treat the quantum motion properly, the total length scale spans orders of magnitude, demanding the development of novel multiscale theoretical approaches tailored for such quantum systems.

#### 4.1.4. Strain effects in growth of semiconductor quantum dots

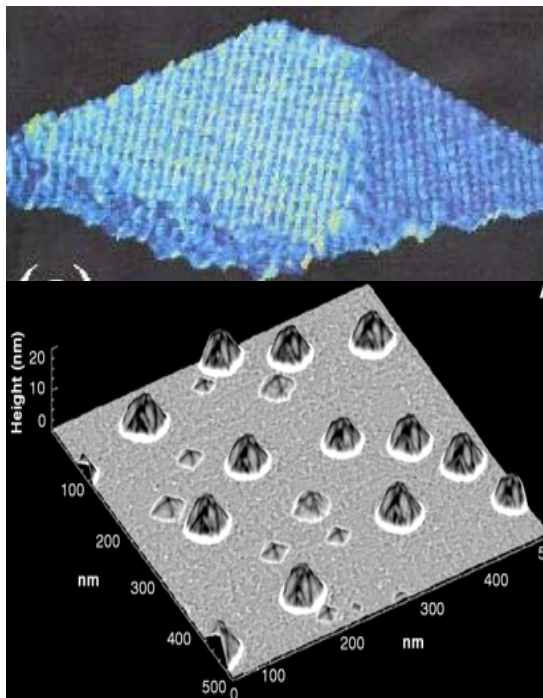


Fig.4. (upper) STM image of a single Ge dot (hut) [1]. (lower) STM image of an array of Ge dots (hut and dome) [61].

Strain-induced self-assembly of strained 3D islands (see, e.g., Ge islands in Fig. 4) in both elemental group-IV and compound III-V semiconductor thin films has been a major area of fundamental research [60]. The growth of faceted quantum dots in lattice-mismatched systems is generally understood on the basis of a competition between the energy cost to create the extra surfaces associated with the dots and the elastic strain energy that can be relaxed by the formation of the dots [62,63]. However, the precise physical origin of their surprising size uniformity remains controversial. Furthermore, good size uniformity has also been achieved recently for 3D metal islands grown on insulator substrates [5], possibly by different mechanisms [64]. Semiconductor islands are usually faceted while metal islands are not, except for some special cases [33]. The former grows generally in the Stranski-Krastanov (SK) mode with wetting layer while the latter may grow in the Volmer-Weber (VW) mode without a wetting layer; the former islands have the same crystal structure as

the substrates while the latter is different. These differences may be significant in determining how to drive and control the self-assembly process.

Another issue in self-assembled 3D islands is to control the nucleation on the surface. In recent experiments, 3D islands were grown on pre-patterned substrates or along steps on a vicinal substrate, resulting in deposition in specified areas and improved island size uniformity. 3D islands grown on patterned compliant substrates display unique novel growth and mechanical properties [65,66]. A challenge in theoretical modeling is to include the effects of strain due to a patterned substrate, where defects (such as dislocations) or embedded particles in the substrate should govern the spatial arrangement of the quantum dots. A preliminary continuum elasticity theory calculation of growth on a strain-patterned substrate with favorable nucleation sites is

shown in Fig. 5. While the dots grow mainly in regions of low mismatch strain, there is also formation of periodic ripples in regions of high mismatch strain. As deposition progresses, materials diffuse to the substrate leading to a regularly ordered array. Numerical simulations can also keep track of the mean stress in the film, which can be monitored in growth experiments. Such new experiments and simulations may open up new opportunities for theoretical studies not only in the field of strained island growth but also in nanomechanics. Another goal is to include the effects of intermixing, and surface segregation. If this is accomplished, the effect of Si capping and the growth of vertically aligned quantum dots can be studied systematically.

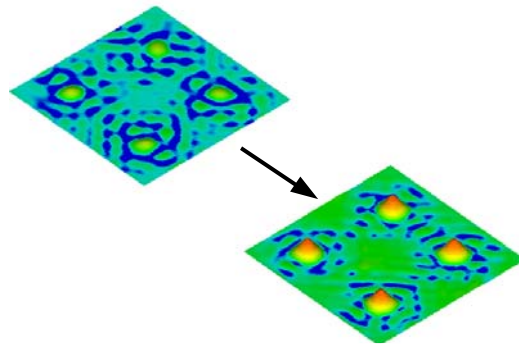


Fig.5: Growth of quantum dots on a strain patterned substrate. The islands nucleate and grow at regions with low mismatch strain [67].

The above proposed continuum modeling and simulation can be greatly enhanced by atomistic calculations of kinetic and thermodynamic parameters. Such a combination will allow the continuum simulations to be carried out with “real” parameters so that the simulations can be made system specific.

While a continuum treatment of elastic energy in quantum dot formation has been quite successful [62-70] (and will be further developed here), the effects of atomistic discreteness are also expected to be important. For examples, wetting layers are just a few layers deep and strain relief mechanisms are manifestly atomistic (e.g., divacancy line or DVL formation); pre-pyramids emerging from wetting layers are only a few layers high. Furthermore, even fully developed faceted pyramids display clear atomic-scale roughness which must influence the strain fields. Also intermixing and inhomogeneities in alloy composition are difficult to treat at the continuum level. Thus, there is strong motivation to implement an atomistic treatment of elasticity. Our efforts in this area will be guided by two goals: (i) to refine continuum treatments of elasticity; (ii) to integrate an atomistic treatment of strain with fully atomistic KMC simulation of growth kinetics. KMC simulations of growth also provide an invaluable tool for treating non-equilibrium aspects of growth kinetics (which are difficult to incorporate precisely in continuum formulations [71,72]). Strong kinetic effects can also exist in the formation of quantum dots, especially at lower temperature [73-75], and they even have been proposed recently for the wetting layer [76]. Thus, it would be very useful to complete atomistic simulations of growth, as well as tailored simulations, to provide insight on non-equilibrium effects as input to continuum modeling.

#### 4.1.5 Quantum size effects in growth of ultrathin metal films

Electronic structure effects, such as quantum size effects (QSE), can have important consequences on the morphology of nanostructures. An intriguing and unexpected feature has recently been discovered in epitaxial growth of metallic nanostructures on semiconductor surfaces. Instead of forming three-dimensional (3D) islands of various heights as commonly observed for nonreactive interfaces, under the right growth conditions the metal atoms can arrange themselves into plateaus or islands of selective heights, with flat tops and steep edges. This unusual behavior was first suggested in the growth of Ag films on GaAs [77]. A similar behavior was also observed in the growth of Ag islands on Si(111), where even at low coverages

the Ag adatoms form isolated islands with a strongly preferred height and flat tops [78]. More recently attention has been focused on the growth of Pb islands/films on Si(111). Fig. 6 shows the STM image of Pb islands on Si(111) of uniform height of about seven layers after deposition of 3 ML at 192 K [79].

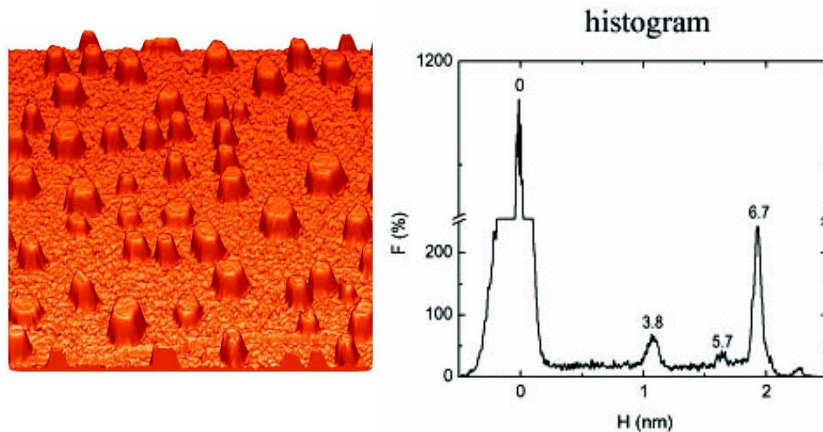


Fig. 6 Left: STM image obtained after deposition of 3 ML of Pb at 192 K on Si(111) showing the uniform height islands with flat tops and steep edges. The scale is  $200 \times 200 \text{ nm}^2$ . Right: A height histogram showing the preferred heights in the image [79].

The formation of these uniform, self-organized atomic structures points to a potentially interesting pathway to prepare functional metallic nanostructures. It is believed that this extra stability of metal films with specific thickness has an electronic origin [58] and can be explained by the “quantum size effect” due to electron confinement [80-82]. The itinerant electrons in metal films are confined in the direction perpendicular to the film surface, resulting in discrete energy levels associated with the so-called quantum-well (QW) states. As the film thickness increases, the energy subbands sequentially cross the Fermi level. Every time a new QW state crosses the Fermi level and gets occupied, we will see a cusp in the system's physical properties. The stability of the film would also be affected due to the variation in the electronic energy.

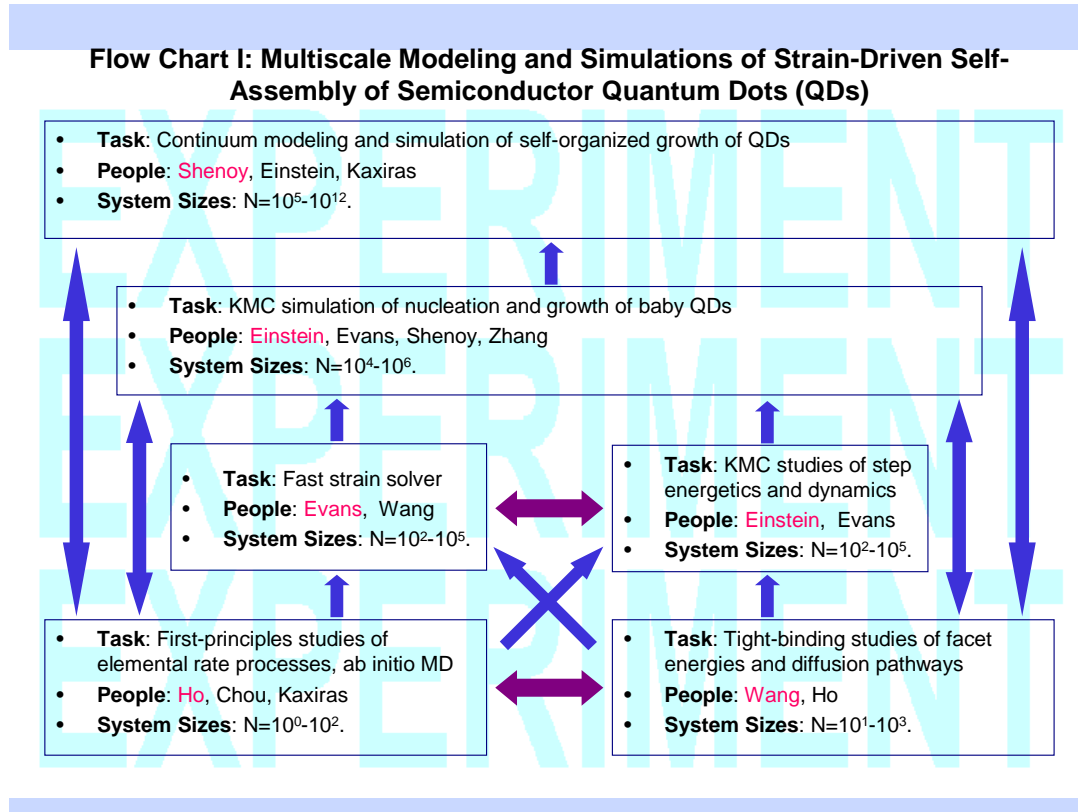
Within the last year or two, a flurry of experimental investigations have uncovered many interesting aspects of the Pb/Si(111) system. An alternating layer and island growth of Pb on Si was observed in real-time *in situ* x-ray studies and attributed to a spontaneous quantum phase separation [26]. Two types of islands having different substrates were reported that displayed an alternating STM image contrast with their thickness [83]. An experimental scheme is developed to construct Pb nanomesas on Si(111) substrates whose thickness can be controlled with atomic-layer precision using an STM triggering step followed by self-driven and self-limiting mass-transfer processes [84]. These new experimental results call for comprehensive theoretical modeling for this intriguing system, using theoretical approaches spanning all the length scales. Details will be given in subsection 4.2.2.

## 4.2 Proposed Work

Our beginning research will be focused on two prototypical problems: one on strain-induced self-assembly of quantum dots in the  $\text{Ge}_x\text{Si}_{1-x}/\text{Si}(100)$  system (section 4.2.1 below) and the other on quantum growth of ultrathin metallic films in the Pb/Si(111) system (section 4.2.2). A great wealth of experimental studies has been performed on these systems, providing a huge pool of data to verify theoretical models to be developed. Studies on these systems will be used to establish the linkages between simulation tools attacking the problems at different length

scales from the different groups on the team. The methodology developed can further be applied to a number of exciting emerging problems outlined in section 4.2.3.

The flow charts shown on this page and page 17 outline the linkages among the different tasks, team numbers, and length scales of the proposed CRT work. These research tasks will be carried out in close collaboration with the experimentalists committed to participate in the CRT. More details of the proposed research will be given in the following three subsections.



The need for multiscale approaches is the underlying scaffold on which the efforts of the various contributors to this proposal will come together. Expertise at the various levels exists within the group, including the quantum mechanical regime at either the density functional (DFT) or tight-binding (TB) level (**Chou, Ho, Kaxiras, Wang, Zhang**), the mesoscopic level (**Einstein, Evans, Zhang**), and the continuum level (**Evans, Shenoy**). Furthermore, some team members (**Kaxiras, Shenoy, Wang, Ho**) have been devoting substantial research efforts to the development of general multiscale methodologies. The class of problems considered here require a tightly integrated approach at the various levels including expertise both on the methods employed and on the physical systems under study.

For the purposes of the work proposed here, both sequential and concurrent multiscale simulations will be needed. Traditional KMC simulations coupled with information on rate processes from first-principles atomistic calculations are an example of the sequential type. While this may be adequate for homoepitaxy, the systems described below involve significantly more complicated situations, particularly where strain is involved. In this sense, it will be necessary to develop further the type of multiscale simulations involving KMC and atomistic results, so that strain is taken into account properly and the relevant rates are computed under the

appropriate strain conditions. KMC results can also be used to determine whether or not all relevant atomistic processes have been included in the picture, by comparison to experiments. This comparison could necessitate a more detailed analysis of the atomistic processes, at either the TB or DFT level.

#### 4.2.1. Strain driven self assembly of quantum dots in the $\text{Ge}_x\text{Si}_{1-x}/\text{Si}(100)$ system

##### 4.2.1.1 First-Principles and Tight-Binding Studies (Ho, Wang, Chou, Kaxiras)

First-principles total-energy calculations can provide detailed accurate information on thermodynamic and kinetic parameters for various elemental rate processes on the surface. However, in these systems, the geometry is complicated enough that a satisfactory exploration of the relevant phase space and transformation pathways can only be accomplished using a combination of tight-binding and first-principles calculations.

*a) Genetic algorithm search for ground state structure of high-indexed GeSi surfaces:* Studies of the high index (10n) and (11n) surfaces can yield information on step formation energies and step-step interactions which are important inputs in KMC and continuum modeling. However, these surfaces are very complex and the ground state structures are largely unknown. Recently, we have extended the genetic algorithm (GA) developed for structural optimization of atomic clusters [85-88] to the problem of surface geometry optimization [89,90]. We will combine the GA with recently developed tight-binding Ge-Si potentials to determine the atomic geometry of high-indexed GeSi surfaces.

*b) Atomistic/electronic calculation of kink formation energies, step formation energies and step-step interaction energies:* The energies of stepped surfaces depend not only on the detailed chemical bonding at the steps, but also on the strain field created by the atomic reconstruction at the steps. We will calculate the step formation and step-step interaction energies using first-principles and tight-binding methods. Elastic effects due to lattice mismatch induced strain will be handled in an efficient manner by using zone-matching schemes to couple quantum mechanical calculations to calculations with empirical potentials [15].

*c) Quantitative determination of kinetic and thermodynamic growth parameters:* The potential-energy landscapes associated with kinetic processes on surfaces are complex because displacement of the surface atoms are coupled to relaxation of the substrate atoms with many degrees of freedom, and involve concerted motion of atoms [30,36,37]. Efficient search engines with accurate tight-binding potentials can be used to investigate the potential-energy landscapes in detail before applying first-principles approaches. In the past several years, we have successfully used this method to study Si adatom and addimer diffusion on the Si(100) surface [36,37,91-94]. We will extend this method to more complex situations such as Ge or Si diffusion and intermixing on Si(100) surfaces in order to understand the kinetics of the GeSi quantum dot formation on Si(100). We will search for the key pathways for Ge and Si atom diffusion on Si(100), at steps, and around kinks.

The results of step and kink formation energies, step-step interactions, pathways and energy barriers for diffusion processes on terraces and at steps and kinks, as well as the influence of strain and Si-Ge intermixing on these quantities, will be used as inputs in the KMC (Section 4.2.1.3) and continuum (section 4.2.1.4) simulations of the morphological evolution of the quantum dots.

#### 4.2.1.2 Atomistic Elasticity and Fast Strain Solvers (Evans, Wang)

The *first application* of our implementation of Cauchy-Born type atomistic treatment of elasticity (“lattice statics”) [95] is to provide inputs to “refined” continuum modeling of elasticity in (4.2.1.4). Such treatments can incorporate complex unit cell crystals and realistic interatomic potentials, and can be integrated with finite element formulations [96] via a quasicontinuum analysis [97]. We will thus be able to derive appropriate macroscopic constitutive relations, and to precisely analyze strain fields and interactions associated with defects such as steps [98,99]. Such analyses will exploit our expertise in atomic level description of Si-Ge systems (4.2.1.1)

The *second application* is development of efficient strain solvers for integration into fully atomistic treatment of film growth. In previous studies of submonolayer islands and adlayers, a Frenkel-Kontorova (FK) type formulation has described equilibrium patterns in binary alloy adlayers [100], and a 2D Green’s function approach has treated initial self-assembly of quantum dots [73]. Previous multilayer studies have just treated simple 2D models using: novel (boundary integral type) Green’s function methods [101]; finite-difference formulations amenable to numerical math methods (e.g., adaptive mesh refinement) [102,103]; and various other local relaxation methods [104].

Our goal of analyzing more realistic 3D models will require very efficient strain solvers. To this end, we are developing a “strain dynamics” technique for fast relaxation which should efficiently recover equilibrium configurations. This work exploits our expertise in MD, but greatly improves on running standard MD [105] to achieve relaxation. Given the complexity of the models, we plan to test and apply this approach first on specific geometries or features (e.g., a single pre-pyramid or pyramid). Other specific analyses will include: development of a FK type approach to treat strain relief in wetting layers (e.g., initiated via DVL’s) and in just-nucleated 2D strained multi-component quantum dots (in 3D systems); development of other efficient strain solvers for growth processes based on local relaxation and perturbation theories.

#### 4.2.1.3 KMC Simulation and Step Dynamics for QD Formation and Structure (Einstein, Evans, Shenoy, Zhang)

Some recent work has already attempted to incorporate efficient atomistic strain solvers into atomistic treatments of growth [73,101,104,105]. E.g., Sander et al. [101] showed that a single atomistic model for strain-mediated growth can provide a unified description of the transition from nucleation-mediated growth of QD’s for large misfit to development of QD’s via the ATG instability at low misfit. However, such studies have been mainly for simplified 2D models.

We will pursue the challenge to develop and implement this approach for more realistic models in 3D. For KMC simulation, key inputs are the hopping rates,  $h = \nu \exp[-E_{\text{act}}/kT]$ , for surface atoms, where the barrier  $E_{\text{act}}$  depends on the local environment (via chemical bonding) and global morphology (via strain),  $\nu$  is the attempt frequency, and  $T$  is the surface temperature. In simplified models,  $E_{\text{act}}$  is artificially separated into two components. Instead, our strain analysis above will provide the total energy for configurations before and after hopping, and with the particle removed. From these energies, we will construct a physically reasonable  $E_{\text{act}}$ .

Our complete atomistic simulations of growth will explore key phenomena in QD formation including: (i) non-equilibrium nucleation and growth of 2D strained islands (cf. [73]); (ii) unconventional [106] nucleation-less formation of 3D Ge or Si-Ge islands on Si(100) [107,108]. For the latter, STM [108] reveals the atomistic details of the barrier-less formation of rounded “pre-pyramids” which eventually become metastable and convert into {105} faceted

pyramids through a first-order transition [69]. Continuum theories incorporating an anisotropic surface energy qualitatively reproduce this behavior [61,62]. Atomistic modeling will examine in detail this noise-induced transition. Such modeling will heavily exploit atomistic input from section 4.2.1.1.

Another important output of atomistic modeling will be to provide direct input to continuum modeling in 4.2.1.4, e.g., on surface mobilities determining equilibrium mass fluxes.

Finally, additional studies will be performed based on step dynamics formulations [109] to explore the dynamics and associated fluctuations of the growing QD's. More generally, we are interested in the effect of strain on step structure and dynamics, as an ability to control step stiffness and step bunching is a valuable tool facilitating the formation of quantum wires (see section 4.2.3 for more details).

#### **4.2.1.4 Continuum Modeling (Shenoy, Einstein, Kaxiras)**

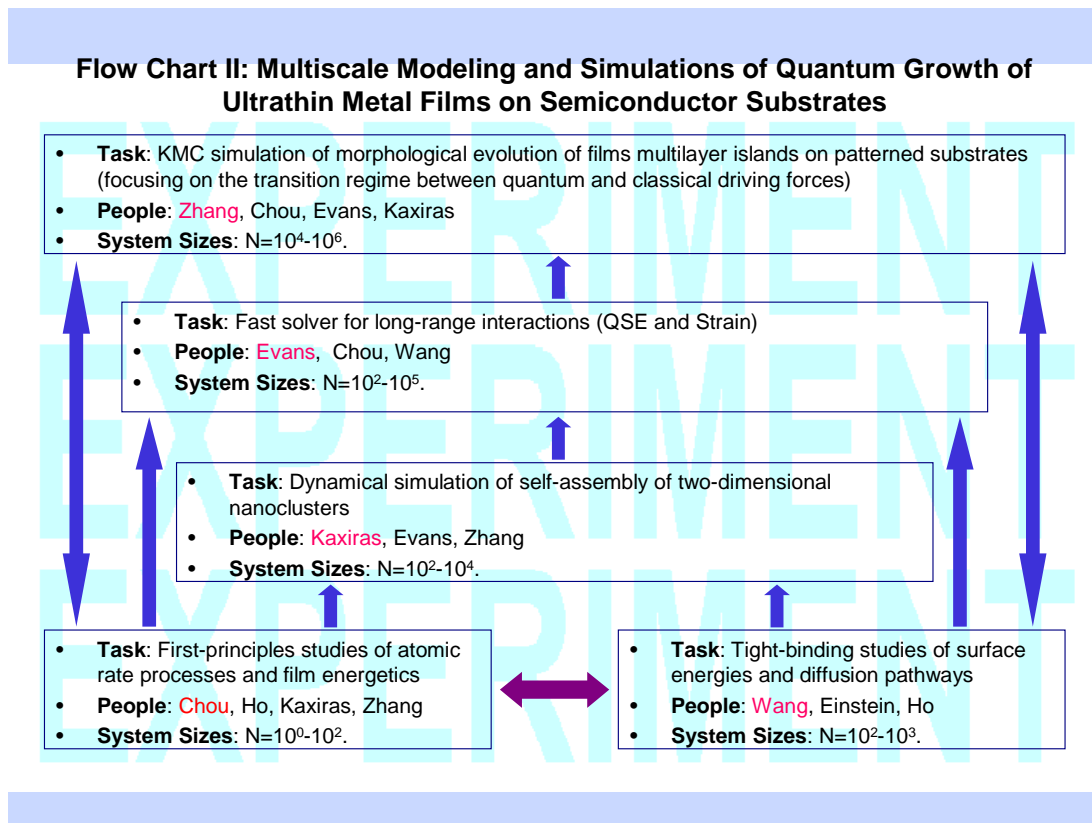
*a) Spatial ordering and interactions of quantum dot arrays:* Ordering of nanostructures can be obtained on patterned substrates using the strain fields of defects such as misfit dislocations, buried wires or mesas or periodically etched grooves. We propose a combined experimental and theoretical effort to understand the nucleation, growth and evolution of quantum dots on patterned substrates. We will use a continuum model including anisotropies in surface energy and kinetic barriers due to surface steps (from section 4.2.1.1) to model growth of quantum dots on these surfaces. Nanostructure growth on such substrates can be influenced significantly by the kinetics of mass transport on surfaces - while there are energetically favorable locations for the islands to nucleate and grow, it is possible that, under some growth conditions, the deposited material would not be able to find the low energy configurations due to kinetic constraints. As deposition progresses, material prefers to diffuse to the substrate leading to a regularly ordered array. Numerical simulations can also keep track of the mean stress in the film, which can be monitored in growth experiments. We will use this method to study ordering and correlations in vertically stacked quantum dot systems.

*b) Compositional effects in nanostructures:* Application of quantum dots as functional elements in nanoelectronics usually requires capping of these structures with a material that is lattice matched to the substrate, so that difference in the material properties would lead to confinement of charge in these nanostructures. In this process, it is important that the dots retain their shapes during and after the capping process. Recent experiments show that, at typical growth temperatures, possible intermixing of the capped Si with Ge in the SiGe quantum dots can lead to reduction of the strain, which results in a change in its shape. Furthermore, it has been observed that a significant amount of Si is able to make its way from the substrate into the Ge quantum dot during growth at elevated temperatures, so that the core of the dot is rich in Si. Understanding these processes requires a study of the coupling of the compositional variations via bulk and surface diffusion to elastic fields in these nanostructures. We propose to address these important phenomena by employing phase-field models that incorporate anisotropies in surface energies, alloy segregation effects, and long-range elastic fields. We will use these simulations to model the stability and evolution of the quantum dots exposed to a capping flux and intermixing with the material from the substrate.

*c) Effect of step-edge barriers on evolution of quantum dots:* Morphological evolution and stability of the alloy nanostructures depend critically on kinetic parameters such as the terrace diffusion constants and the step-edge barriers encountered by different atomic species. Just as in the case of single component surfaces, experiments on the relaxation of periodic ripples,

produced for example by sputtering the surface with inert gas atoms can be used to extract these material properties. While relaxation of material surfaces above the roughening temperature is described by the curvature based linear theory, first developed by Mullins, the evolution of ripples below the roughening temperature are governed by dynamics of surface steps, which are highly non-linear. We have recently developed a variational method to solve these non-linear equations and have applied it to study the evolution of ripples on semiconductor surfaces [110]. We propose to apply this technique to study the effect of step-edge barriers on the evolution of quantum dot arrays. With input on kinetic parameters from atomistic simulations described in subsection 4.2.1.1, we will use our continuum model to study ways of achieving better order in the quantum dot arrays.

#### 4.2.2 Ultrathin metal films on semiconductor substrates



##### 4.2.2.1 First-Principles and Tight-Binding Calculations (Chou, Wang, Ho, Kaxiras, Zhang)

In order to better understand the quantum-size effects of metal thin films and to better control the growth of these metallic nanostructures, we will first employ state-of-the-art computational methods to study a series of systems. Working closely with the experimentalists, we will first address the electronic properties of the metal overlayers on semiconductors and the resulting stability issues by first-principles density-functional calculations. The goal is to predict the orientation and thickness dependence of the stability threshold of flat-top metal islands of a few *s-p* metals such as Al, Mg, and Ag. Our first results for Pb thin films have confirmed the oscillatory quantum size effects exhibited in the surface energy and work function [111]. In

contrast to the commonly used free-electron-like models, it was found that a quantitative description of these quantum size effects required full consideration of the crystal band structure.

Tight-binding electronic and total-energy calculations will be used as a complementary method to first-principles calculations to deal with larger systems (several hundreds to several thousands atoms), and with more complex situations where structures or transformation pathways need to be explored. We have recently developed a set of environment-dependent tight-binding potentials for the Pb/Si systems [22]. Such tight-binding potentials will be further improved so that they can be used to study the Pb/Si interface structures and to explore the diffusion pathways and energy landscapes for the growth of Pb nanoislands on the Si(111) surface.

*a) Interface structure optimization:* While the concept of “quantum size effect” is very appealing and can partially account for some of the intriguing experimental observations, previous theoretical studies of such effects used only simplified models such as 1-D square potential wells [21,58] or free-standing metallic films [111]. It is known from experiments that the stability of the metallic islands can also be affected by the structure of the metal/semiconductor interface [20,26,112]. Our recent study shows that the electronic structure of the Pb film is very sensitive to the atomistic relaxation at the Pb/Si interface [22]. Depending on the position of the Fermi level and the atomic arrangements at the interface, different substrates may have different effects, leading to different stability conditions. Using a combination of tight-binding molecular dynamics and the genetic algorithm (GA), together with information from X-ray experiments (Tringides), we will perform structure optimization for Pb wetting layers and films on different Si substrates. We will also calculate the electronic structures and make comparisons with the results of STM and photoemission experiments in order to understand the electronic properties (particularly band bending) and the charge transfer at the interfaces. The optimized structures from the tight-binding studies will be used in first-principles calculations for detailed studies of the energetics and electronic properties of Pb films on the Si(111) surface.

*b) Quantitative determination of kinetic and thermodynamic growth parameters:* We will search for the key pathways for Pb atom diffusion on Si(111), on the Pb wetting layers, and on the top of the Pb islands using methods described in section 4.2.1.1. The resulting parameters will be used as inputs in the KMC simulations. Some of the plausible pathways will also be passed to first-principles calculations for more detailed studies.

*c) Generalization to other related systems:* What we learn from the Pb/Si(111) system will be expanded to other metal/semiconductor systems, e.g., the closely-related system Pb/Ge(111) offers convincing experimental evidence for quantum growth (bilayer growth, beating, and unusually persistent quantum behavior as a function of film thickness).

#### **4.2.2.2 KMC Studies of Growth Incorporating Long-Range Interactions (Evans, Zhang, Chou, Kaxiras, Wang)**

Atomistic modeling and kinetic Monte Carlo (KMC) simulation of the growth of flat-top metal islands with sharply defined and highly preferred heights on semiconductors is an outstanding challenge. It is complicated by the presence of long-range interactions related to quantum size effects (QSE), and also to strain effects. Usually, strain interactions dominate film growth, but in these systems QSE-related interactions primarily control the growth behavior. This phenomenon is intrinsically a kinetic effect, so it is necessary for modeling to incorporate

appropriate determination of activation barriers for diffusion of metal atoms across flat regions, on the steep sides of islands, onto the tops, etc. Extending the modeling strategy incorporating strain effects in heteroepitaxy without QSE, the hopping rates for these systems will be written in the form  $h=v \exp[-(E_0 + E_{\text{bond}} - \delta E_{\text{strain}} + \delta E_{\text{QSE}})/kT]$ . Here,  $E_{\text{bond}}$  accounts for chemical interactions;  $\delta E_{\text{strain}}$  reflects the change in strain energy upon hopping (and is determined as discussed in 4.2.1.1), with the added feature that detailed analysis of the structure of the wetting layer will likely be critical in its analysis. Finally,  $\delta E_{\text{QSE}}$  reflects QSE effects. It is non-local, as it must reflect preference for certain film thicknesses, and some insight into its form has been explored in our preliminary studies [59]. Essential inputs to the KMC modeling regarding the form of  $\delta E_{\text{QSE}}$  will come from the electronic and atomistic calculations described in 4.2.2.1.

Specific experimental input to our modeling will come from STM studies of “triggered” growth kinetics of individual layers on top of the islands [84,113], and from multistage deposition studies [114]. These studies probe key barriers controlling growth and reveal important details of growth mechanisms and morphologies which must be matched by effective modeling. Another fascinating aspect of kinetics in these systems is the evolution of these metastable states with flat selected-thickness islands towards the equilibrium 3D island state (usually following annealing) [13]. Such studies provide insight into kinetic pathways and barriers quite distinct from the above-mentioned growth studies, and thus will also provide a valuable testing ground for our modeling.

#### **4.2.3. Emerging opportunities for fundamental research with immense technological significance.**

The modeling and simulation capacities acquired in the first two subsections using the prototype model systems of  $\text{Ge}_x\text{Si}_{1-x}/\text{Si}(100)$  and  $\text{Pb}/\text{Si}(111)$  and  $\text{Pb}/\text{Ge}(111)$  will enable a broad spectrum of challenging issues to be addressed by the materials science community, including this CRT. Here, we choose three distinct classes of problems as our longer-term research objectives, 1) growth of carbon nanostructures on  $\text{Si}(100)$ , 2) fabrication of quantum wires on stepped surfaces, and 3) quantum engineering of magnetic nanoplatelets. The solution of each class of the problems will directly benefit from the collective knowledge and conceptual advances from both of the above subsections, and promises to have important impact in technological applications.

##### **4.2.3.1. Growth of high-strain nanostructures on Si (Kaxiras, Evans, Ho, Shenoy).**

The  $\text{Ge}_x\text{Si}_{1-x}/\text{Si}(100)$  system is representative of the variety of issues that arise when a relatively small amount of strain exists between the substrate and the overgrown layers, ranging up to 4% for pure Ge. Strain is generally viewed as an agent that introduces difficulties into the growth process. However, this feature could also be used as an agent that allows for more efficient manipulation of surface morphology. In this sense, it would be desirable to have a large range of strain values, which ideally could be controlled as a continuous variable. In fact, the introduction of carbon on Si surfaces acts as such an agent [115,116], since carbon has a covalent radius that differs from that of Si by as much as 30%. Interestingly, the presence of carbon leads to unusual reconstructions on the Si surface, which can be exploited as templates for the growth of novel nanostructures. These effects are brought about by relatively small amounts of subsurface carbon atoms at the Si surface. It is unclear how exactly carbon is incorporated, what the relative stability of different carbon-induced reconstructions is, and how the range of reconstructions depends on the amount of carbon present and the conditions of deposition [117].

Here we propose to explore the possibilities of large strain in heteroepitaxial systems, in order to determine conditions that can give rise to new and interesting superstructures, such as dots or wires, driven by the morphological features that arise from the strain release processes. We will use carbon on Si surfaces as the prototypical large-strain system and methodically investigate the mechanism of incorporation, the strain effects on surface reconstruction, the strain release mechanism such as creation of dislocations, microvoids, etc. The goal will be to determine how, by taking advantage of large strain, one could control the creation of features on the surface with desired dimensions and distribution, and how such features can be exploited for the growth of superstructures with predictable properties.

#### **4.2.3.2. Fabrication of quantum wires and quantum wire superlattices.**

Our objective here is to devise kinetic pathways for fabrication of ideal one-dimensional nanostructures as testing grounds for fundamental physics and technological applications.

For fundamental physics, such as exploration of non-Fermi liquid behavior in 1D transport, we will explore the possibility of growing ideal 1D metal wires on a hydrogen-passivated diamond (100)-2x1 substrate. The choice of a diamond substrate is based on its superb thermal and mechanical properties, and, more importantly, its wide band gap and the corresponding minimal participation of the substrate in electrical transport, making the metal wires potentially truly one-dimensional. The metal elements will include Ti, which has a large cohesive energy (over 7 eV) at the C(100) surface, and Al, which is a better conductor in bulk form. Here, the straightness of the wires will be secured via selective removal of unwanted hydrogen using techniques such as scanning probe-based nanolithography. Our preliminary DFT studies have shown that the Ti wire is strictly one dimensional with regard to electrical transport. We will utilize the multiscale simulation methods developed using the prototype model systems to define the precise growth conditions at which ideal arrays of such parallel 1D quantum wires can be fabricated via self-assembly of Ti or Al atoms deposited on a patterned diamond (100) surface. A comparative study will also be made on Si(100), to contrast the different degrees of participation of the substrates in electrical transport (**Chou, Evans, Zhang**).

An even more intriguing objective is to devise kinetic pathways for possible formation of quantum wires and quantum wire superlattices via step decoration on various vicinal substrates. Here, the preparation of the stepped substrates with extended straight steps will benefit from what we have learned about step dynamics and, in certain cases, bunching. Furthermore, the interaction of adatoms with pre-existing steps will need to be understood at the first-principles level, so as to capture all the important atomic processes, in many cases concerted in nature, determined by both the local bonding and the electronic confinement effect. For the case of quantum wire superlattices consisting of alternating magnetic and nonmagnetic elements, maximum energy for phase segregation along the steps is desired, which can be drawn from chemical, mechanical/strain, and magnetic driving forces. Recent experimental efforts along this line have seen limited success [7]. Our goal is to explore theoretically the diverse growth conditions and compositional parameters for the best chance to fabricate such dream nanostructures (**Einstein, Kaxiras, Zhang**).

#### **4.2.3.3. Quantum engineering of magnetic nanoplatelets (Zhang, Chou, Evans, Kaxiras, Wang).**

Empowered by the multiscale modeling and simulation approaches achieved in the previous two subsections, in particular by the major conceptual advances in understanding the

delicate interplay between various thermodynamic driving forces (QSE, strain, interface energetics, ...) and kinetic factors, we can set out to tackle more intriguing scientific problems that otherwise would be too demanding computationally. One representative system to address is the formation, via quantum engineering, of magnetic (Co or Fe) nanoplatelets on patterned Si(111)-7x7 substrates. Recently, working with our collaborators on the experimental side, we (Zhang and Shih) have managed to form on Si(111) large arrays of metal nanoclusters that are identical in size and spatially uniform. This advance itself is a beautiful example of delicate control of the interplay between growth kinetics and thermodynamics, utilizing the “attractive basin” idea proposed by a team member (Kaxiras in Ref. 53). An example for the case of Al is shown in Fig. 7(a). Even more remarkably, when atoms of a magnetic element such as Fe or Co are deposited onto the Si(111)-7x7 substrate pre-decorated with the Al magic nanoclusters, through fine tuning of the growth conditions, they manage to form equilateral triangular islands that possess singular lateral and vertical sizes. The lateral sizes are selected by the 7x7 unit cells, while the vertical height, of precisely two atomic layers, is likely the result of enhanced confinement of the electron motion due to the passivation of the Si surface by the Al nanoclusters. Preliminary studies have revealed that these Co nanoplatelets possess a magnetic blocking temperature proportional to the number of the Co atoms in the islands, namely, the whole platelets can be magnetized with strong magnetization anisotropy [29]. To fully understand how the magnetic nanoplatelets are formed, we will need to assemble all the tools and concepts emphasized so far. These include various energetic and kinetic factors, the competition between quantum size effects, strain, and local chemical bonding at the interfaces; the possible existence of sub-atomic scale corrugation and intermixing, and interaction between different nanoplatelets. The microscopic origin of the unusual magnetic properties of the nanoplatelets poses an additional challenge and motivation, and its elucidation must rely on precise knowledge of the atomic arrangement in the complex system. Success in this research direction will have an important impact on efforts to integrate ultrahigh density information storage and information processing on single silicon chips.

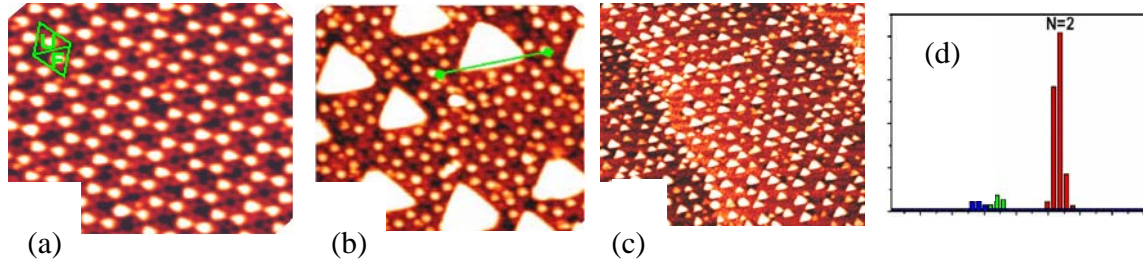


Fig. 7. Magnetic nanoplatelets formed on a pre-patterned Si(111)-7x7 substrate. (a) Formation of magic Al nanoclusters on Si(111)-7x7, in which every half faulted or unfaulted unit cell collects precisely six Al atoms. (b) Co atoms deposited on the Al<sub>6</sub>/Si(111)-7x7 substrate form nanoplatelets that possess equilateral triangular geometry with unique orientation, as well as quantized lateral and vertical dimensions. (c) With proper control of the growth conditions, the Co nanoplatelets predominantly have the same lateral sizes as shown in (d), where N is the number of the 7x7 unit cells along an edge of the triangular islands.

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